REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	2/21/96	Final Report 12/16/94-12/14/95

4. TITLE AND SUBTITLE
Study of Anion-Molecule Reaction Dynamics with Time-Dependent Photoelectron Spectroscopy

5. FUNDING NUMBERS
61103D
3484/US

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AFOSR-TR-96
0122

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Air Force Office of Scientific Research/NL
Chemistry and Materials Science Research
110 Duncan Avenue, Suite B115
Bolling Air Force Base, DC 20332-0001

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
F49620-95-1-0078

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution is unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

A novel femtosecond time-resolved experiment has been set up to probe the photodissociation dynamics of negative ions and negative ion clusters. First results for the photodissociation of I_2^- have been obtained recently.

19960320 041

14. SUBJECT TERMS
negative ions, photodissociation, femtosecond photoelectron spectroscopy

15. NUMBER OF PAGES
3

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLISSIFIED	UNCLASSIFIED	UNCLASSIFIED	UNLIMITED (SAR)

FINAL TECHNICAL REPORT

TITLE: Study of Anion-Molecule Reaction Dynamics with Time-Dependent Photoelectron Spectroscopy

PRINCIPAL INVESTIGATOR: Daniel M. Neumark

DATE: December 16, 1994 - December 14, 1995

GRANT NO: F4960-95-1-0078

Abstract:

A novel femtosecond time-resolved experiment has been set up to probe the photodissociation dynamics of negative ions and negative ion clusters. First results for the photodissociation of I_2^- have been obtained recently.

Final Technical Report:

DURIP funds were used to purchase a Ti:sapphire femtosecond laser system (Model #CPA-1000 MPS) from Clark MRX Inc., at a cost of $165,600. The Ti:sapphire femtosecond laser system is being used on a new experiment designed to study the photodissociation dynamics of negative ions and negative ion clusters on a femtosecond time scale. The novel feature of this experiment is that photoelectron spectroscopy is used to probe the dynamics of the dissociating anion in real-time. We have very recently obtained our first results on I_2^- photodissociation.

The principle of the experiment can be best understood by specifically considering its application to I_2^-:

$$I_2^- \xrightarrow[h\nu_1]{} I_2^{-*} \xrightarrow[\Delta t]{} I\cdot\cdot I^- \xrightarrow[h\nu_2]{} I_2 + e^-,\ I + I + e^-$$

This is a pump-probe experiment using two femtosecond laser pulses. The pump pulse ($h\nu_1$, 780 nm) excites mass-selected I_2^- from the ground $X\ ^2\Sigma_u^+$ state to the repulsive $^2\Pi_{u(3/2)}$ state. The I_2^- begins to dissociate, and after a variable time delay Δt, the dissociating ions are photodetached with the probe pulse ($h\nu_2$, 260 nm). We then measure the photoelectron spectrum, thereby mapping out the dissociating wave packet onto the well-known I_2 potential energy surfaces.

At short times, one expects to see a transient associated with the dissociating I_2^- molecule, and at longer times, the I^- photoelectron spectrum should become more prominent as dissociation occurs. This is precisely what has been observed in our first measurements on I_2^-. However, the time scale for dissociation appears to be several hundred fsec longer than predicted by our simulations that use the currently accepted potential energy curves for I_2^-, indicating that these potentials may require significant modification. This issue will be addressed in further experiments and theoretical work.